

Modelling the redox conditions inside a failed nuclear waste container

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Abstract

The recommended approach for the long term management of used nuclear fuel in Canada is adaptive phased management which includes the option of isolation of the used fuel in centralized containment in a deep geological repository. In the unexpected event of container failure two corrosion fronts would exist within the groundwater-flooded container: one on the fuel surface driven by radiolytic oxidants, and a second one on the carbon steel surface sustained by water reduction and producing the potential redox scavengers, Fe^{2+} and H_2 . As illustrated in Fig. 1, a complex series of homogeneous solution and heterogeneous surface reactions will determine the redox conditions within the failed container and, hence, the fuel corrosion/radionuclide release rates.

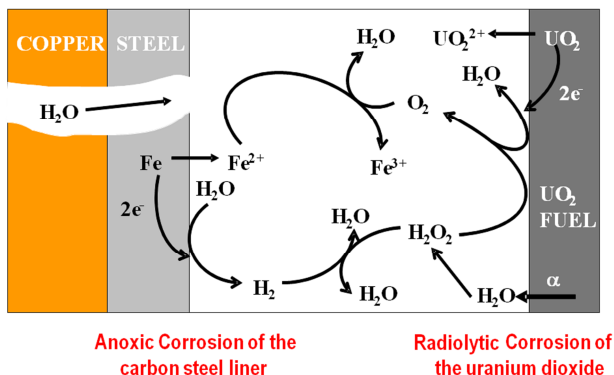


Fig 1. Schematic showing the corrosion scenario inside a failed nuclear waste container.

A model has been developed to investigate the influence of steel corrosion products on the α -radiolytic corrosion of spent nuclear fuel. As illustrated in Fig. 2, the model includes: (I) the generation of H_2O_2 by α -radiolysis of water; (II) the oxidative dissolution (corrosion) of UO_2 supported by H_2O_2 on both the UO_2 surface and noble metal particles; (III) the reduction of oxidized $\text{U}^{\text{V}}/\text{U}^{\text{VI}}$ catalyzed by H_2 oxidation on noble metal particles; (IV) the scavenging of H_2O_2 in homogeneous solution by reaction with Fe^{2+} ; (V) the decomposition of H_2O_2 to O_2 and H_2O ; (VI) the anoxic corrosion of steel container.

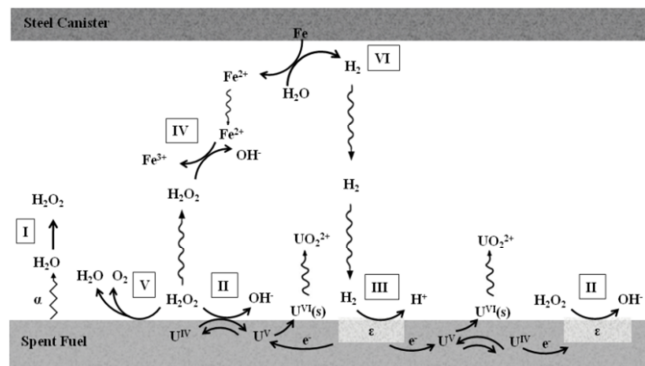


Fig. 2. Reactions considered in the model.

The model is based on finite element methods and developed using COMSOL Multiphysics. A one-dimensional model is used to investigate the influence of Fe^{2+} and H_2 on fuel corrosion. The calculation is based on the α -dose rates for an aged CANDU fuel bundle with a burnup of 220 MWh kgU^{-1} . The dominant redox control agent is found to be H_2 . The ability of Fe^{2+} to scavenge H_2O_2 by the Fenton reaction has only a minor influence on the fuel corrosion process. Critical H_2 concentrations, the $[\text{H}_2]_{\text{crit}}$ required to completely suppress fuel corrosion, are calculated as a function of α -dose rate for various spent fuel ages. A two-dimensional model is also under development to investigate the efficiency of $\text{H}_2/\text{Fe}^{2+}$ in suppressing fuel corrosion at locations within fractures in fuel pellets, locations to which the transport of $\text{H}_2/\text{Fe}^{2+}$ will be limited.

Keywords: Nuclear materials, Modelling studies, Uranium dioxide, Corrosion.